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1 SHEET

COMPLETE SPECIFICATION

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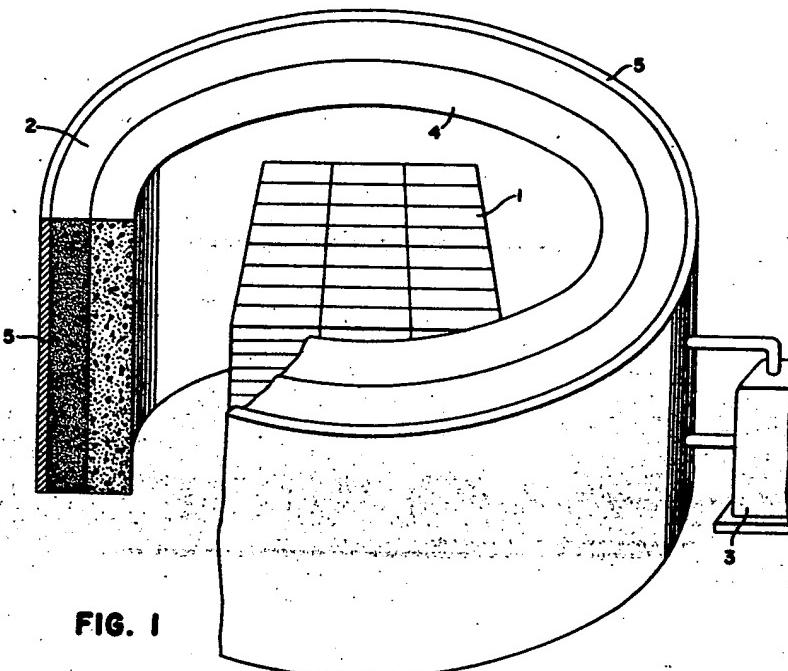


FIG. 1

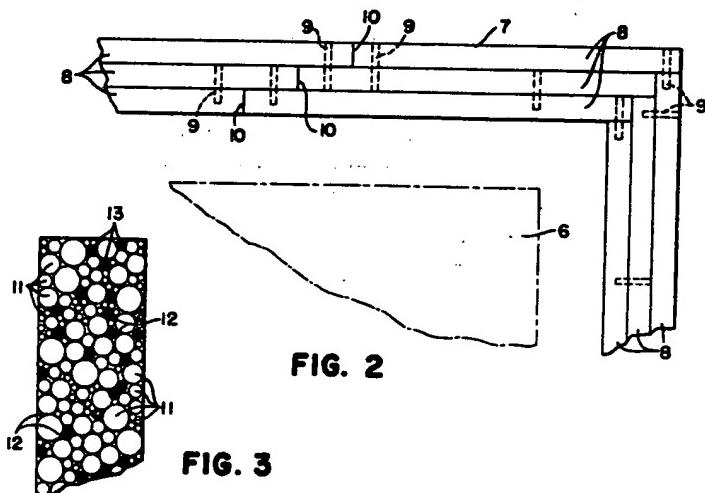


FIG. 2

FIG. 3

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PATENT SPECIFICATION

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DRAWINGS ATTACHED.

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COMPLETE SPECIFICATION.

Composition of Matter Useful as a Neutron Attenuant.

We, THE GOODYEAR TIRE & RUBBER COMPANY, a corporation organized under the Laws of the State of Ohio, United States of America, with offices at 1144 East Market Street, Akron, Ohio, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:

This invention relates to a composition of matter useful as a neutron shield or attenuant for a nuclear reactor or other neutron source.

Recent improvements in the technology of reactor engineering have made possible the production of small and medium sized atomic reactors for use as the principal energy source of propulsion systems for vehicles, aircraft, ships and the like. Such reactors utilize a fissionable mass ordinarily having present therein a substantial proportion of uranium 235. In the process of fission large quantities of heat are produced, which heat may be utilized, by means of conventional heat exchange devices, to drive an engine to propel a number of types of vehicles. During the process of fission there are, in addition, large amounts of various radiations emitted which, if allowed to propagate unimpeded, have an extremely harmful effect on human organisms, oft times resulting in death if sufficient quantities impinge on body tissue. One of the most difficult forms of radiation to contain is the neutron since it has no electrical charge and is of relatively high velocity when emitted during the fission process and therefore difficult to control. It has thus been a problem in the past to provide a light-weight, compact

shield for use in mobile installations which acts to slow down and absorb neutrons for the protection of personal who must operate within the immediate area of a reactor. It is, of course, well known that the attenuation or slowing down properties of any material insofar as neutrons are concerned, are directly proportional to the so-called hydrogen density of the material. In general, the hydrogen density is proportional to the number of hydrogen atoms per unit volume present in the shielding material. In early prior art devices water was commonly utilized for neutron shielding since it had a reasonably good hydrogen density. Unfortunately it presented problems because the water had to be contained in a heavy bulky tank and maintained in a pure state to avoid radioactive contamination. Advances in shielding techniques led to the use of polyethylene which has a much better hydrogen density than water and, since it is a solid under ordinary conditions, requires no tank or other container. However, the erection of a shield made of polyethylene requires the fabrication of the shield out of many individual slabs, which is not only time consuming but tedious. Furthermore, the polyethylene slabs have a tendency to buckle and crack during installation and use, as well as a tendency to develop voids during the manufacturing stage, so that there is always the danger of a loss in shielding properties in a slab constructed shield. Other materials, such as butyl rubber, which has a hydrogen density comparable to polyethylene, have been considered for use in a neutron shield but unfortunately materials such as butyl rubber have a relatively low damage thresh-

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old and hence a tendency to disintegrate under radiation. Thus their use is limited to low neutron flux areas. Butyl rubber also presents some difficulty in effecting a cure to provide a self-sustaining structure. Thus, the problem of providing an easily constructed and dependable shield for high flux areas has remained.

It is a principal object of the present invention to provide a composition of matter useful as a neutron attenuator or shield which can be easily and economically constructed. It is a further object of the present invention to provide a shield material which has a hydrogen density which approaches that of polyethylene when fabricated into a shield without the disadvantages of a solid slab construction. It is a still further object of the present invention to provide a shielding material for an atomic reactor which can absorb physical shock and impact with a minimum risk of fracture. An additional object of the present invention is to provide a pourable castable neutron attenuant.

As previously mentioned, the provision of an adequate neutron shield for propulsion type reactors and other sources of neutron radiation has long presented difficult problems. While polyethylene and similar materials have a high hydrogen density, they also possess certain physical and mechanical characteristics which render fabrication of an adequate shield difficult and time consuming as well as present substantial risk of mechanical failure. Such shields must be built of a series of large slabs which must in some way be fastened together in a manner as to avoid what might be termed neutron leak areas. Furthermore, due to the particular type of construction necessary to utilize slab materials, the shields so constructed may require greater volumes in a given vehicle than is strictly necessary for shielding if all available volume is utilized. Hence, under certain circumstances the total reactor volume including its shield is unnecessarily larger. Furthermore, it is difficult to erect a shield to enclose an irregular area due to the fact that slab materials are being used. These problems are satisfactorily met by the present invention which, in general, provides a castable or mouldable shield material having a hydrogen density comparable to solid polyethylene. The castable or mouldable feature of the shield of the present invention possesses obvious advantages in that the volume occupied by the reactor and its shield can be minimized in terms of unused shielded volume. Furthermore, it is possible to erect the shield by means of in situ casting to enclose irregular areas or shapes such as auxiliarly heat exchange tubes.

This invention consists in a composition of matter useful as a neutron attenuant,

comprising a major proportion of solid particles of polyethylene or polypropylene and a minor proportion of a pourable, settable organic binder. The composition of this invention might be termed an organic concrete. As will later appear, it has further been discovered that by the proper choice of a number of sizes and amounts of aggregates, a castable, mouldable shield material is provided which possesses a hydrogen density only a few percent less than polyethylene or polypropylene alone, but which has the distinct advantages of being castable or mouldable in character as well as possessing a higher capability of absorbing mechanical shock.

The invention will be further described, by way of example, with reference to the accompanying drawings, of which:—

Fig. 1 is a partially sectioned view of a nuclear reactor enclosed by a shield, the principal neutron absorbing portion of which is made according to the present invention;

Fig. 2 is a partial plan view of a nuclear reactor surrounded by a slab constructed shield made according to the prior art; and

Fig. 3 is a cross sectional view of a portion of the shield made in accordance with the present invention.

Referring to Fig. 1, a reactor 1 of the classic pile type is schematically shown surrounded by a moulded shield 2. If the reactor is to be used for propulsion purposes, it is usually equipped with some type of heat exchange apparatus designated generally by the numeral 3. The shield 2, which in the form shown comprises a major proportion of finely powdered polyethylene held in a minor proportion of a pourable, settable organic binder, functions to slow down and stop neutrons emitted by the pile 1, but may not necessarily provide adequate shielding for other types of penetrating radiation such as gamma rays. It is understood therefore, that secondary biological shields 4 and 5 comprising heavy materials such as concrete and lead to provide gamma shielding must necessarily be provided in many cases. The essentially continuous or homogenous character of the shield 2 can be appreciated by an examination of Fig. 1 and the absence of joints, bolts, or fastening devices and similar undesirable non-radiation absorbing elements usually needed in slab type shields clearly seen.

It is understood, of course, that in certain installations, such as nuclear powered aircraft, the shield may entirely enclose the area occupied by the crew or other personnel rather than the reactor itself. In this way substantial savings in shield thickness may be effected due to the greater separation possible between the reactor and shield and hence the lower neutron fluxes encountered. In addition, it is thus possible to minimize

the problem of forming the shield components about heat exchange piping, control apparatus, and similar objects.

Fig. 2 illustrates the situation which exists with a prior art slab-type polyethylene shield, the usual auxiliary shields being omitted for the sake of clarity. The reactor 6 is surrounded by a slab polyethylene shield designated generally by the numeral 7 which comprises a plurality of polyethylene slabs 8. The slabs may be connected to each other by fastening means 9 which may be bolts, dowls, or similar devices and which preferably are compatible with polyethylene from the standpoint of physical characteristics such as thermal expansion. In the construction of a slab type shield 7 it is necessary to either stagger or lap the joints 10 between the individual slabs in order to avoid a neutron leak, i.e. a continuous void in the neutron shielding material. It can readily be appreciated that if the slabs do not fit perfectly flush together at the joints, it is necessary to increase the thickness of the shield in order to compensate for this loss of shielding property. Thus, although polyethylene and similar materials have high hydrogen densities and, therefore, are very effective neutron decelerators, the slab construction fails to take full advantage of the nuclear characteristics of the material and it may oft times be necessary to build a shield of more than minimum shielding thickness in order to provide the necessary protection. Furthermore, as can be seen from the type of stucture illustrated in Fig. 2, the difficulty in building a shield to enclose an irregular area or to fit around piping, etc., as may be required in certain propulsion applications, is evident. It may be concluded therefore that, although the castable shield material of the present invention may not possess quite as high a hydrogen density as polyethylene, it may in many applications nonetheless be more efficient in terms of actual shield volume required. In addition it is more easily installed by casting in a conveient mould.

Fig. 3 illustrates a cross section of a portion of another form of a neutron shield made according to the present invention. The shield comprises a major proportion of a plurality of discretely sized, preferably substantially spherical polyethylene particles 11 held in substantially close-packed relationship in a minor proportion of a binder 12 which, as will appear, may be a variety of materials. It has been found generally essential that the polyethylene particles 11 be chosen to be of at least three discrete sizes which do not approach each other in particle diameter but vary from size to size by a factor of approximately 10, and preferably by a factor of 100, in order to secure high packing factors. It is understood, of

course, that some variation in particle diameter within each size is to be expected due to inaccuracies inherent in the manufacturing process. However, no diameter should vary from the average by any substantial amount which would interfere with the packing. In some circumstances, it may be desirable to utilize a more open-packed arrangement where flexibility of shield is of importance. In general, however, the close-packed arrangement is preferred where shielding efficiency is of primary concern. While irregularly shaped particles may be used with good results, substantially maximized packing and fluidity is obtained using generally spherical particles. It has further been found that in order to minimize the void space and, since the binder is generally of substantially lower hydrogen density, thus maximize the hydrogen density of the material, the proper ratio of the amounts of each sized particle for maximum density is preferably $1:v_1:v_1^2:v_1^3\dots = V_1:V_2:V_3:V_4\dots$ where v_1 is the void space of any single component (the void space, which is the ratio of the voids to the total volume, being generally independent of particle size), V is the amount of material, and the subscripts refer in inverse order to decreasing particle size, i.e. V_1 is the amount of the largest particle size and V_4 is the amount of the smallest, the particle sizes varying by a factor of 10.

The binder 12 may be of a variety of materials, it having been found that a copolymer of butadiene and methacrylic acid, which will later be described, provides an elastomeric material with sufficient physical strength for construction of the shield and at the same time one which possesses a reasonably good hydrogen density. While polyethylene particles are preferred as the aggregate, polypropylene may be utilized without a decrease in hydrogen density. However, polyethylene being in large commercial supply and of reasonably low cost, is the preferred material.

Pure high density polyethylene has a hydrogen density of approximately 0.138, while low density polyethylene has a hydrogen density of about 0.134. A shield material comprising a curable or vulcanizable copolymer of butadiene and methacrylic acid including powdered, but not discretely sized, polyethylene was found to have a hydrogen density of approximately 0.118, or only 14% less than pure polyethylene. A shield material made according to the present invention, utilizing the same copolymer and a two component polyethylene aggregate having particle sizes of 0.36 centimeters and 150 microns, respectively, exhibits a hydrogen density of 0.128 gram per cc or only about 7% below pure polyethylene. According to the present

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invention a shield material using particles of polyethylene of at least three discrete sizes, having a ratio of smallest to largest particle of approximately 0.0005, results in an even higher hydrogen density, approaching that of pure polyethylene and similar materials. It is preferred, of course, that in this form of shield at least three and preferably a greater number of discrete particle sizes be used in order to secure maximum hydrogen densities.

Since the copolymer of butadiene and methacrylic acid has a hydrogen density of only about 0.104, it has been found generally to be desirable when utilizing such a material as the binder to minimize the void fraction of the polyethylene in order to reduce the amount of polymer necessary to fill the voids and provide the necessary fluid characteristics by proportioning the polyethylene to assure the nearest approach to a close-packed arrangement. Flow properties were found to be imparted to a mixture of polyethylene particles when only a slight excess (for example 1 to 5 per cent) of copolymer over the amount necessary to fill the voids is present. It is obvious, of course, that for any given choice of powder or discrete particle sizes, the void space can either be calculated or determined empirically and, hence, the amount of binder necessary can be easily determined. Decreasing the void space by increasing the number of discrete particle sizes results in two distinct advantages: first, the hydrogen density is increased by reducing the lower hydrogen density liquid polymer to a minimum, and secondly, the decreased void space reduces the amount of liquid polymer necessary to impart fluidity to the system. Furthermore, the use of generally spheroidal particles, either in a homogenous powder or the distinct sizes, tends to minimize the effective surface area which must be wet by the binder in order to provide suitable flow characteristics. Because it is pourable and castable, the shielding material avoids the difficulties found in a polyethylene slab type shield in that fitting around the shield enclosure and reactor rigging is no longer a problem since the shield material can be poured in a mould about the reactor and any auxiliary piping or equipment and vulcanized in place. As previously mentioned, where added flexibility of shield is desired and less shielding efficiency can be tolerated, the close-packed arrangement may be replaced by an open-packed cubical or even more widely dispersed particle disposition.

A pourable binder of the above type may be made by conventional polymerization techniques employing mixtures of 100 parts by weight butadiene with varying amounts (preferably from 9 to 10 parts by weight) of methacrylic acid in the presence of relatively

high levels, for instance about 12 parts by weight, of modifiers such as mercaptans of bis xanthogens. More specific description of other monomers which may be employed and of the methods for preparing these polymers will be found in United States Patent 2,395,017 and in an article by H. P. Brown in "Rubber Chemistry and Technology," Volume 30, No. 5, December, 1957, pages 1347 et seq. These liquid polymers are suitably cured through the use of various cross linking agents but preferably through the use of an epoxide such as the epoxide marketed as Epon 562. The amount of curing agent will vary depending upon the specific material used, it having been found that from 18 to 20 parts of Epon 562 per 100 parts of polymers provides a particularly effective curing system. Although elevated temperatures of up to about 200° F. may be used to accelerate the process and reduce cure time, such a binder has the advantage that it will cure or set at room temperatures to a satisfactory strength.

While the binder and polyethylene may be combined in numerous ways as by pouring the components into a container and agitating them to assure substantially homogeneous distribution, it is preferable that when utilizing various sizes of polyethylene particles they be added simultaneously in order to avoid a non-homogenous particle disposition and, hence, to ensure a nearest approach to the desired packing.

While minor amounts of a variety of materials may be added to the elastomers thus produced in order to enhance the physical characteristics or chemical resistance thereof, it is usually desirable to avoid materials such as zinc oxide since they contribute very little to the shielding efficiency but present a problem since they tend to become radioactive in the presence of neutron radiation, necessitating further shielding. In addition, materials such as polymethyl methacrylate, which degrade under irradiation and release gases during the process, are also preferably avoided.

Utilizing a castable shield material utilizing the discretely sized particles, which has a hydrogen density of about 0.128, with some contribution in shielding qualities due to the presence of carbon but only minor contributions by the small amounts of sulfur and oxygen contained thereon, a macroscopic neutron cross section of about 0.108 cm^{-1} for neutrons at 10 mev energy can be realized, while a shield utilising polyethylene powder provides a cross section of about 0.104 cm^{-1} . A polyethylene slab shield of about six centimeters thickness, if of voidless character, will provide fifty per cent shielding according to the formula $I = I_0 e^{-E_t}$ where I_0 is the initial or unshielded neutron

- intensity and I is the neutron intensity after attenuation by the shield, E is the cross section, and t the thickness. In contrast, a castable shield using the discretely sized particles will provide the same shielding with an increase in shield thickness of only about four-tenths of a centimeter and the powder type shield an increase of only about sixty-five one-hundredths of a centimeter. Such an increased thickness may be more than compensated for by the castable nature of the material which permits savings in total shielded reactor volume as compared to a slab structure as well as substantial reduction of the risk of voids.
- As mentioned, other binders may be used to secure a castable, mouldable, high hydrogen density shield. In instances where a personnel shield is desired and the incident neutron flux is not unduly high, butyl rubber or polyisobutylene, which have a hydrogen density comparable to polyethylene, may be used as the binder and plasticizer without serious difficulties. However, effecting a satisfactory cure presents some problems with butyl rubber. Since butyl rubber (a copolymer of a major proportion of an iso-olefin such as isobutylene with a minor proportion of an open-chain conjugated diolefin such as isoprene) has a hydrogen density very similar to polyethylene, the resulting shield not only is as effective as polyethylene in terms of neutron attenuation but retains the mouldable and castable characteristics and their inherent advantages. In addition, certain high melting point waxes and asphaltic materials may satisfactorily be used as a binder material although the elastomers referred to above possess certain advantages due to their resilience. Such materials may be heated to achieve a fluid state in order to facilitate addition of the polyethylene and neutron absorbing materials and then, after pouring in situ at the reactor, be allowed to cool and set. Of course, as with the previously described shield materials, a shield may be prefabricated in sections if desired and then transported to and erected at the site.
- In order to provide for the efficient absorption of thermal neutrons, i.e. neutrons which have been slowed down or attenuated by the shield material, it is desirable to incorporate in a homogenous dispersion throughout the shield minor proportions, such as up to 10 per cent by weight, of high neutron-absorptive materials, designated by numeral 13 in Fig. 3. These materials preferably contain boron, lithium, or cadmium which have thermal neutron absorption cross-sections of at least 60 barns. In addition, other elements having a high thermal neutron absorption cross section such as samarium, gadolinium, europium, dysprosium, erbium, and iridium may be used although cost and availability present some problem. In addition, certain of these materials emit secondary gamma rays after capturing a neutron and therefore necessitate increased auxiliary shield, while this effect is substantially less when the preferred materials such as lithium are used. It is understood, of course, that for certain applications these materials may be omitted and the polyethylene-binder mixture used alone as an attenuator. As previously mentioned, secondary shields to provide for efficient absorption of gamma and X-rays and similar dangerous radiations may be provided either inside or outside the neutron shield or in both locations as desired and needed.
- While certain representative embodiments and details have been shown for the purpose of illustrating the invention, it will be apparent to those skilled in this art that various changes and modifications may be made therein without departing from the scope of the invention as defined in the appended claims.
- WHAT WE CLAIM IS:—**
1. A composition of matter useful as a neutron attenuant, comprising a major proportion of solid particles of polyethylene or polypropylene and a minor proportion of a pourable, settable organic binder.
 2. A composition according to Claim 1 in which the binder is a vulcanizable elastomeric binder.
 3. A composition according to Claim 1 or 2 in which the polyethylene or polypropylene particles are substantially spheroidal.
 4. A composition according to any of Claims 1 to 3 in which the polyethylene or polypropylene is dispersed in a substantially close-packed arrangement.
 5. A composition according to any of Claims 1 to 4 in which the polyethylene or polypropylene particles are provided in a plurality of substantially discrete sizes.
 6. A composition according to any of Claims 1 to 5 in which the polyethylene or polypropylene particles are provided in at least three substantially discrete sizes.
 7. A composition according to Claim 5 or 6 in which the particle sizes vary from size to size by a factor of approximately 10.
 8. A composition according to Claim 7 in which the particle sizes vary from size to size by a factor of approximately 100.
 9. A composition according to Claim 7 or 8 in which each size is present in an amount determined by the formula $1 : v_1 : v_1 : v_1^2 : v_1^3 \dots = V_1 : V_2 : V_3 : V_4 \dots$ where v_1 is the void space (as hereinbefore defined) of a single sized component, V is the amount of said component, and the subscripts refer in inverse order to decreas-

- ing particle size, the particle sizes varying by a factor of 10.
10. A composition according to any of the preceding claims which contains a minor proportion of a material having a thermal neutron absorption cross section of at least 60 barns.
11. A composition according to Claim 10 in which the neutron absorptive material comprises a major proportion of lithium, boron, or cadmium.
12. A neutron shield comprising a com-
- position according to any of the preceding claims.
13. A composition of matter in accordance with Claim 1 and substantially as described and set forth hereinbefore.
14. A neutron shield in accordance with Claim 12 and substantially as described with reference to, and as shown in, either Fig. 1 or Fig. 3 of the accompanying drawings.

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